Effect of Thermal Undulations on the Bending Elasticity and Spontaneous Curvature of Fluid Membranes

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Abstract

We amplify previous arguments why mean curvature should be used as measure of integration in calculating the effective bending rigidity of fluid membranes subjected to a weak background curvature. The stiffening of the membrane by its fluctuations, recently derived for spherical shapes, is recovered for cylindrical curvature. Employing curvilinear coordinates, we then discuss stiffening for arbitrary shapes, confirm that the elastic modulus of Gaussian curvature is not renormalized in the presence of fluctuations, and show for the first time that any spontaneous curvature also remains unchanged.

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Introduction

The fluid membranes of red blood cells and giant vesicles in water undergo microscopically visible thermal shape fluctuations [1]. The thermal undulations lead to orientational decorrelation on a scale characterized by de Gennes' persistence length [2]. About fifteen years ago, one of us proposed that the decrease of correlation should be accompanied by a softening of the membrane. Specifically, the effective bending rigidity κ' was predicted to obey

$$\kappa' \;=\; \kappa - \alpha \frac{kT}{8\pi} \log M \;, \tag{1}$$

where κ is the bare bending rigidity and M the number of molecules making up the membrane. The numerical prefactor was found to be $\alpha = 1$ in the first derivation [3] and two later ones intended to support this result [4, 5]. In all three papers mean curvature was assumed to be the statistical measure. However, serious mistakes were made in the calculations. Peliti and Leibler, whose result was the first to be published [6], and all other authors [7, 8, 9, 10, 11, 12] rederived eq. (1) for the renormalized bending rigidity, but with the prefactor $\alpha = 3$. They took normal displacement of the membrane from its equilibrium shape as measure of integration. Only recently the problem was reconsidered [13] when the mistakes were noted in the calculations employing mean curvature. In addition to correcting the previous calculation for the sphere [4], a new calculation was done for the almost flat fluctuating membrane. It is based on a local bending mode expansion called hat model [14]. In both cases it was found that fluid membranes should be stiffened, not softened, by their thermal undulations. The form of the relationship (1) is maintained, while the numerical prefactor changes sign, becoming $\alpha = -1$. However, it has be mentioned that a few years ago Gompper and Kroll found a membrane softening in Monte Carlo simulations of fluctuating vesicles [15].

In the present work we deal once more with the problem of selecting the correct statistical measure, taking advantage of the analogies between polymers in two-dimensional space and fluid membranes. Subsequently, we consider a particularly transparent situation, cylindrical curvature, to rederive the effective bending rigidity in this case. Employing curvilinear coordinates, we then find the stiffening, i.e. the validity of eq. (1) with $\alpha = -1$, for more general deformations of the initially flat membrane. In the same tensor notation, we confirm that the elastic modulus of Gaussian curvature is not affected by thermal undulations and show for the first time that this also holds for any spontaneous curvature of the membrane.

Formulation of the problem

The bending energy of a fluid membrane may be expressed by the Hamiltonian

$$H = \int \mathrm{d}^2\sigma \ \sqrt{g} \left(\frac{1}{2}\kappa J^2 - \kappa c_s J + \bar{\kappa} K \right) \ , \tag{2}$$

where κ and $\bar{\kappa}$ are the bending moduli and c_s is the spontaneous curvature. The terms in parentheses represent the surface energy density and $\sqrt{g} \ \mathrm{d}^2\sigma$ is the local surface element of the membrane. Let $\vec{X}(\bar{\sigma})$, $\bar{\sigma} = (\sigma^1, \sigma^2)$, be a parametrization of the membrane surface with the contravariant coordinates σ^1 and σ^2. Then $\mathrm{d}^2\sigma = \mathrm{d}\sigma^1 \mathrm{d}\sigma^2$ and $g = \det g_{ij}$, where

$$g_{ij} = \partial_i \vec{X} \cdot \partial_j \vec{X} \ , \quad \text{with} \quad \partial_i \vec{X} = \partial \vec{X}/\partial \sigma^i \ , \quad i, j = 1, 2 \ ,$$

are the coefficients of the first fundamental form. J and K denote trace and determinant of the extrinsic curvature tensor $J^i_j = g^{ik} J_{kj}$, respectively, where

$$J_{ij} = \vec{N} \cdot \partial_{ij} \vec{X} \ , \quad i, j = 1, 2$$

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are the coefficients of the second fundamental form, $\vec{N}$ being (in the case of a closed surface) the outward unit normal vector to the surface. In other words,

$$J = c_1 + c_2 \quad \text{and} \quad K = c_1 \cdot c_2 \ ,$$

are twice the mean curvature and the Gaussian curvature, respectively, with c_1 and c_2 being the principal curvatures. Note that with this convention J is negative for spheres and cylinders.

The concept of an effective rigidity can be understood as follows: Starting from a basically flat, weakly fluctuating piece of membrane, we are interested in the free energy Γ needed to deform the piece into a fluctuating shape of nonvanishing background curvature J. It turns out that the effect of the short wavelength fluctuations upon the elastic behaviour on large scales can be absorbed into the material parameters of the bending Hamiltonian (2). Accordingly, the contribution to Γ from the first term in (2) may be written as

$$\Gamma \ = \ \frac{1}{2}\kappa' \int J^2 \sqrt{g} \ \mathrm{d}^2\sigma \ . \tag{3}$$

Here the curvature and the surface element refer to the nonfluctuating background and $\kappa' \neq \kappa$ is the effective bending rigidity.

As is common in shape calculations, the membrane is assumed to be unstretchable. The free energy Γ refers to a background surface of fixed area. The associated real membrane area is slightly larger because of the bending fluctuations. Any shrinkage of base area resulting from the fluctuations can be formally avoided by introducing a (fictitious) reservoir of basically flat membrane area. Alternatively, Γ can be taken to refer to the shrunken base area. Such a shrinkage does not affect the bending energy of the base if it is done by an isotropic scale transformation, as happens automatically in the case of a freely fluctuating spherical vesicle. Fortunately, these subtleties seem to be irrelevant in practice.

The free energy of bending, Γ, can be obtained in two different, but equivalent ways. One may "freeze" the undulations in an instantaneous configuration and then calculate the bending energy of the rippled membrane at fixed undulation amplitudes. The squared fluctuation mode amplitudes are averaged only afterwards.

Alternatively, one may start from the free energy of the fluctuating flat membrane and investigate how it changes when the membrane is subjected to a background curvature $J \neq 0$. The fluctuation dependent part of the bending energy Γ is now entropic, resulting from the effect of J on the mean square amplitudes of the undulation modes.

In both cases we are faced with the problem of choosing the right measure of integration. The undulations as expressed by the measure have to be kept constant when the membrane with "frozen" undulations is deformed in the large. Correspondingly, the entropic part of the free energy of background deformations has to be calculated from the change of the mode mean square amplitudes in terms of the measure. We will expand now on some of the arguments why mean curvature, or any quantity proportional to it, is a correct measure of integration. The scale of the measure is of no interest because we are dealing with differences, not absolute values of entropy.

Considerations on the measure of integration

A heuristic argument for choosing mean curvature as statistical measure is the fact that it can be regarded as the physical strain of the fluid membrane, much like curvature in the case of a (stiff) polymer. Accordingly, the superposition of two deformations, e.g. uniform background curvature and a (sinusoidal) ripple, begins with adding the strains as they are in the absence of the other deformation. The simultaneous presence of both deformations will produce coupling terms in the ef-

fective total bending energy that are quadratic in both the background curvature and the ripple amplitude. Employing the strain as statistical measure has the particular attraction of avoiding spurious coupling terms. They arise from a change of the ripple amplitude in terms of the strain when other measures are used such as normal displacement from the background or reorientation of the layer normal.

From a technical point of view, mean curvature can serve as measure of integration only if its local values determine the shape of the polymer or membrane, respectively. A one–to–one relationship between shape and local curvatures (which may be molecular) clearly exists for the polymer in the plane, a simplified model system frequently considered in the following. For membranes, a similar invertible mapping is particularly transparent in the hat model which expands the fluctuations of the infinite planar membrane into local mean curvature modes[13]. There are only minor restrictions to the local mean curvature fluctuations in other geometries, such as periodic boundary conditions, cylinders or spheres (see below). However, any (small) membrane deformation compatible with the boundary condition can be completely specified either by local normal displacements or mean curvatures. In all these cases Gaussian curvature fluctuates together with mean curvature, but the former is completely slaved by the latter because of the boundary conditions. The general possibility of invertible mappings between shape and mean curvature, including local area conservation and lateral flow, has been discussed elsewhere [5, 13].

Finally, there is a cogent reason to select (mean) curvature as statistical measure for polymers in 2D or fluid membranes. It stems from the fact that in both systems every local strain fluctuates independently of all the others (apart from the minor restrictions just mentioned). As a result, the total fluctuation entropy is the sum of the equal entropies of all the molecular (or other local) fluctuations. Independence

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and equality imply that the entropy S_q of a sinusoidal (mean) curvature mode on a flat background does not depend on its wave vector q. This is because for all $q \neq 0$ the molecules are in the same way homogeneously distributed over the phases of the sine wave. The q–independence of S_q indicates that mean curvature is a suitable measure of integration since the mean square curvature amplitudes also do not depend on q. Let us add as a remark that the absence of a wave vector dependent Jacobian can perhaps be regarded as characteristic of a correct measure.

The condition of the mode entropy being independent of q is violated if the entropy is calculated in terms of other statistical measures. Taking normal displacement, one generally derives

$$S_q = \frac{1}{2}k \log q^4 \ + \ const. \ ,$$

from the variation with $1/q^4$ of the mean square displacement amplitude of a sinusoidal fluctuation mode. The q–dependent part of S_q is half as strong if normal displacement is replaced by the tilt angle of of the polymer or membrane with respect to the background. Evidently, neither normal displacement nor tilt angle is a correct measure of integration. It should be noted that in many statistical–mechanical calculations the choice of the measure is irrelevant. However, it matters whenever the coupling of two bending deformations is considered as will be demonstrated below.

After these general statements let us address particular model systems to deal with the statistical mechanics of both polymers in a plane and fluid membranes. They will confirm the claim that mean curvature is a correct measure of integration for fluid membranes. Examples of the invertible mapping between membrane shape and local mean curvature will also be given, including the restrictions on the latter.

We first have a look at the comparatively simple problem of a polymer chain fluctuating in a plane [17]. The configurational space of the polymer is swept completely

by varying the angles between adjacent links. Disregarding fluctuations of link length, one may speak of local curvatures instead of angles. Each joint between subsequent links has its own free energy arising from its curvature fluctuations. Although the change of a particular angle rotates the two sides of the polymer chain with respect to each other, it does not affect their free energies. We assume here that the polymer is almost straight. In a continuum description such a polymer is equivalent to a (thin) rod or wire.

Using local curvature as the measure of integration in dealing with fluctuating polymer chains is not in conflict with the fundamental measure which for mass points is translation. Local curvature is a shorthand notation for relative displacement if, e.g., each joint of the polymer chain is regarded as a mass point. Then a (small) normal displacement of point $n + 1$ from the tangent to the polymer defined by the two preceding points is proportional to the curvature at point n. In contrast, the absolute normal displacement of a nearly straight fluctuating polymer from a base line is not a suitable statistical measure. It implies a freedom of motion for each individual mass point which actually is suppressed by cohesion and bending stiffness. The remaining freedom is fully accounted for by the curvature.

The polymer allows a simple demonstration of the effect of a wrong statistical measure on the curvature amplitude of a ripple. We consider a sinusoidal wave on a polymer with a uniform background curvature $-1/r$, the negative sign indicating a downward bend. A sketch of the situation is given in Fig. 1. Let us keep all normal displacements $\nu(s)$ fixed, s being the arc length of the background, while we bend the polymer as a whole. This amounts to using the displacement measure. The procedure results in a weakening of the ripple curvature because the total curvature changes from $\mathrm{d}^2\nu/\mathrm{d}s^2$ to $-1/r + \mathrm{d}^2\nu/\mathrm{d}s^2 + \nu/r^2$ up to linear order in ν, with the two terms constituting the ripple curvature being of opposite sign. Clearly, the change of the ripple curvature amplitude is physically not acceptable so that normal

displacement does not appear to be the correct statistical measure.

Continuing the two–dimensional polymer into the third space dimension results in a bent membrane with zero curvature in the new direction. The similarity of such a membrane with the polymer in a plane strongly suggests that some kind of curvature rather than displacement from a fixed base should be the right measure of integration for fluid membranes.

In order to extend the preceding argument to membranes that can be bent not only in a single direction let us consider a fluid membrane made of mass points. A particularly simple situation arises for the almost flat, weakly fluctuating membrane: Like the polymer, the idealized fluid membrane can be build up by adding the constituent mass points one by one. For easy statistics, we may think of a (local) lattice of mass points, quadratic or hexagonal, which is formed row after row. A new molecule continuing a straight sequence completes a plaquette comprising a certain number of preexisting neighbours. If the number is large enough the plaquette can be used to define the mean curvature at its center. Five molecules is the minimum; they may be thought to form a centered square, as is illustrated in Fig. 2. The height of the last molecule added, relative to a suitably chosen tangent plane, finalizes the value of the curvature J. On a quadratic lattice, J may be taken (in lowest order) to be the sum of the (normal) curvatures of the two diagonals of the plaquette. Let us emphasize, that it is not the relative height of the final molecule that gives a parametrization of the phase space of the particle in the middle of the plaquette, but the simultaneously varying mean curvature. This quantity is invariant with respect to the direction along which rows are formed one by one. The tangent plane may be replaced by the base plane, shifted to the local height, if the fluctuations are weak enough to leave the membrane almost flat. Obviously, the last simplification is possible in the example of Fig. 2.

Regardless of the measure of integration, it is convenient to describe the thermal undulations superimposed on a background shape of given J and K in terms of normal displacement $\nu(\bar{\sigma})$. To linear order, ν and the associated extra curvature $\xi = J' - J$ are related through

$$\xi = [J^2 - 2K + D^2]\,\nu\,,\tag{4}$$

D^2 being the Laplace–Beltrami operator (see eq. (7) below and subsequent definitions). The difference ξ is equivalent to J as statistical measure.

Undulation modes can be defined as eigenfunctions of the operator represented by the bracketed expression. For surfaces of uniform background curvature, such as planes, cylinders and spheres, this ensures that for each mode the functions $\xi(\bar{\sigma})$ and $\nu(\bar{\sigma})$ are proportional to each other in a first order approximation, the factor of proportionality depending on the wave number. The transformation (4) can be inverted whenever for a given undulation mode the operator in brackets does not vanish or affect the membrane area linearly in the mode amplitude. Well–known examples of such exceptions are the three $l = 1$ modes of the fluctuating sphere which actually represent translations and the $l = 0$ mode which requires area changes of the unstretchable membrane. With periodic boundary conditions the only lost mode is uniform mean curvature ($q = 0$) as it is incompatible with the boundary conditions. In the case of the cylinder two long–wavelength curvature modes are lost because they represent translations normal to the cylinder axis. Periodic boundary conditions along the axis and area conservation suppress two other mean curvature modes. The loss of modes implies that the local mean curvatures are not totally free to fluctuate independently of each other. However, these restrictions seem irrelevant in the present context for membranes comprising large numbers of molecules.

Basic formulas

In order to calculate the bending Hamiltonian (2) for a fluctuating membrane, we first present expressions for the mean curvatures and the surface elements of membrane shapes lying close to a given background shape. The latter is also referred to as the projected shape.

If $\vec{X}(\bar{\sigma})$, $\bar{\sigma} = (\sigma^1, \sigma^2)$, is a parametrization of the background configuration, any slightly deviating configuration $\vec{X}'(\bar{\sigma})$ may be represented in normal gauge [7, 10, 18] by

$$\vec{X}'(\bar{\sigma}) = \vec{X}(\bar{\sigma}) + \nu(\bar{\sigma})\vec{N}(\bar{\sigma}) , \tag{5}$$

$\vec{N}(\bar{\sigma})$ being the local outward unit normal vector to $\vec{X}(\bar{\sigma})$. From (5) we find that up to quadratic order in ν the surface element of the deformed membrane obeys (cf.[7, 8, 18, 19, 20])

$$\sqrt{g'} = \sqrt{g}\left(1 - \nu J + \frac{1}{2}(D\nu)^2 + K\nu^2\right) . \tag{6}$$

In the same approximation the curvature J' becomes

$$J' = J + (D^2 + J^2 - 2K)\,\nu - \frac{1}{2}\,J\,(D\nu)^2$$

$$+ J^{ij}\,D_i\nu D_j\nu + (J^3 - 3JK)\,\nu^2 + 2J^{ij}\,\nu D_i D_j\nu +$$

$$+ \nu\,D_n\nu\,g^{ij}(D_j J_i^n + J\,\Gamma_{ij}^n) , \tag{7}$$

Here, g_{ij} and J_{ij} are the coefficients of the first and second fundamental forms of the background surface, respectively. Γ_{ij}^l are the Gauss–Christoffel symbols of the second kind derived from g_{ij}. D_i is the covariant derivative being defined as $D_i\nu = \partial_i\nu = \partial\nu/\partial\sigma^i$, and $D_i D_j\nu = \partial_{ij}\nu - \Gamma_{ij}^k\partial_k\nu$. $D^2 = g^{ij}D_i D_j$ is the Laplace–Beltrami operator and we use the abbreviation $(D\nu)^2 = D^i\nu\,D_i\nu$. The variation of the Gaussian curvature K need not to be considered in (2) thanks to the Gauss–Bonnet theorem [21].

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Renormalization of κ for the cylinder via frozen mode amplitudes

Let us now derive the effective bending rigidity for cylindrical background curvature in the "frozen mode" approach. We first write down reduced versions of (6) and (7) that apply to this special case. Using geodesic coordinates $x = r \cdot \varphi$ and z to parametrize the cylinder, where φ and z are usual polar coordinates, we find with $J = -1/r$:

$$\sqrt{g'} = 1 + \frac{1}{r}\nu + \frac{1}{2}(\nabla\nu)^2 + O(\nu^3) \tag{8}$$

and

$$\begin{aligned} J' = &-\frac{1}{r} + \frac{1}{r^2}\nu + \Delta\nu + \frac{1}{2}\frac{1}{r}(\nabla\nu)^2 \\ &-\frac{1}{r}(\partial_x\nu)^2 - 2\frac{1}{r}\nu\partial_{xx}\nu - \frac{1}{r^3}\nu^2 + O(\nu^3) \ . \end{aligned} \tag{9}$$

The linear part of J' in (9) leads to the transformation (4) and its inversion for the cylinder,

$$(1/r^2 + \Delta)\nu = \xi \ \Leftrightarrow \ \nu = \Delta^{-1}(1 + 1/r^2\Delta^{-1})^{-1}\xi \ , \tag{10}$$

where Δ is the usual Laplacian for a flat base. Obviously, the thermal undulations on a cylinder can be expanded, like those on a flat background, into sinusoidal waves which at the same time are curvature and normal displacement modes.

We will consider a quadratic piece of membrane subjected to a weak cylindrical background bend and satisfying periodic boundary conditions. Alternatively, the membrane may be closed to form a cylinder so that the edge of the square equals the circumference in length. In the displacement representation the sine and cosine deformation modes read

$$\nu = a_{\vec{q}}\sin(\vec{q}\,\vec{r}) \quad \text{and} \quad \nu = b_{\vec{q}}\cos(\vec{q}\,\vec{r}) \ , \tag{11}$$

where $\vec{r} = (x, z)$ and $\vec{q} = (q_x, q_z)$ with $q_x = 2\pi n/L$ and $q_z = 2\pi m/L$. The numbers n and m are running from $-L/(2a)$ to $L/(2a)$, but only over half the $\vec{q}$ plane. The constant mode $(n, m) = (0, 0)$ is excluded. L denotes the size of the background shape and a is the molecular distance in the membrane. The operator acting on ν in (10) is $(1/r^2 - \vec{q}\,^2)$ for a given mode.

In order to renormalize the bending rigidity let us first consider the effect of a single sinusoidal mode. A straightforward calculation starting from (8) and (9) and involving a partial integration then results in the Hamiltonian

$$\int \frac{1}{2}\kappa J'^2 \sqrt{g'} \mathrm{d}x\mathrm{d}z =$$
$$\frac{1}{2}\kappa \left[\frac{1}{r^2} + \frac{1}{2}(1/r^2 - \vec{q}\,^2)^2 a_{\vec{q}}^2 + \frac{1}{2}\frac{1}{r^2}(-\frac{1}{2}q_x^2 + \frac{3}{2}q_z^2 - \frac{1}{r^2})a_{\vec{q}}^2 \right] A_{base}$$

$$(12)$$

where $A_{base} = L^2$ denotes the area of the nonfluctuating background shape. Terms linear in ν do not occur as they vanish on integration. Of the three terms in the brackets, the second one equals the mean value of ξ^2, thus giving the regular bending energy of the mode. Note that if the curvature measure applies, the curvature difference ξ is kept constant in the "frozen mode" approach when the membrane is deformed. Accordingly, the third term is due to the coupling of the mode curvatures to cylindrical curvature.

The statistical averages of the squared mode amplitudes are obtained from the equipartition theorem,

$$\langle a_{\vec{q}}^2 \rangle = \langle b_{\vec{q}}^2 \rangle = \frac{2\ kT}{A_{base}\kappa(1/r^2 - \vec{q}\,^2)^2}\ .$$

$$(13)$$

Inserting (13) in (12) and absorbing the third term of (12) in the first leads to

$$\left\langle \int \frac{1}{2}\kappa J'^2 \sqrt{g'}\mathrm{d}x\mathrm{d}z \right\rangle = \frac{1}{2}\kappa \left(1 + \frac{-\frac{1}{2}q_x^2 + \frac{3}{2}q_z^2 - 1/r^2}{(1/r^2 - \vec{q}\,^2)^2} \frac{kT}{A_{base}\kappa} \right) \cdot \frac{1}{r^2} A_{base} + \frac{1}{2}kT\ . (14)$$

Adding up the contributions of all fluctuation modes to the renormalization of κ yields to lowest order in kT/κ

$$\kappa' \;=\; \kappa \left(1 + \sum_{\vec{q}} \frac{-q_x^2 + 3q_z^2}{(\vec{q}^{\,2})^2} \frac{kT}{A_{base}\kappa} \right) \;, \tag{15}$$

where $1/r^2$ is omitted because of $q > 1/r$. The sum is assumed to be small as compared to unity. It does not contain $(n, m) = (0, 0)$ and some seemingly divergent low wave vector modes. The upper limits of n and m are large numbers. Replacing the sum by the double integral

$$\sum_{\vec{q}} \;\longrightarrow\; \frac{A_{base}}{8\pi^2} \int_{2\pi/L}^{\pi/a} \mathrm{d}^2\vec{q} \;,$$

we arrive with good accuracy at

$$\kappa' \;=\; \kappa + \frac{kT}{8\pi} \log M \;, \tag{16}$$

M being the number of molecules in the membrane (or half this number for the bilayer). Evidently, we have recovered the result derived previously for the fluctuating spherical vesicle, i.e. eq. (1) with $\alpha = -1$.

The effect of thermal undulations on the bending rigidity of fluid membranes was attributed to a coupling between the mode curvatures and the background curvature. To confirm the role of coupling, we may for a moment compensate the uniform background curvature by a spontaneous curvature. For the cylinder this means $c_s = -1/r$ and $J' - c_s = \xi$. Accordingly, the surface energy density is simply $\frac{1}{2}\kappa\xi^2 = \frac{1}{2}\kappa \left(\left(\frac{1}{r^2} + \Delta \right) \nu \right)^2$ up to quadratic order in ξ. The additional quadratic terms arising in the case of interest, $c_s = 0$, can therefore be regarded as coupling terms that give rise to a renormalization of the bending rigidity.

For a direct understanding of the coupling let us consider two special types of ripples, those exactly parallel or orthogonal to the cylindrical background curvature.

Parallel ripple: When the ripple is parallel we can use the analogy to a polymer in a plane. This makes it attractive to use a more direct approach than the general

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calculation based on $\nu(x,y)$. Starting from a straight and stiff polymer or a wire, we impose on it a periodic deformation

$$\xi(s) \; = \; c_q \; \sin(q\,s) \;,$$

where ξ is curvature, s arc length and q wave vector. The undulation diminishes the projected or background length of the wire. Alternatively, it increases the total arc length L' of the wire (when connected to a reservoir) at fixed projected length L. In both cases the ratio of the two lengths is

$$L'/L \; = \; 1 + \frac{1}{4}\frac{\langle c_q^2 \rangle}{q^2} \tag{17}$$

to lowest order in ξ.

A frozen bending mode of the polymer is similar to a wire with a permanent wave. Any sinusoidal bending deformation allows a given angle difference between the ends of the wire to be spread over L' instead of L. This reduces the associated bending energy density by $(L/L')^2$ and the total bending energy by L/L' as compared to a polymer or wire without waviness. The analogous ratio for parallel ripples in membranes is A/A', where A' and A are the real and projected areas, respectively. Absorbing the effect into a renormalized bending rigidity, we have $\kappa' = \kappa \, A/A'$. This agrees exactly with what we obtain from (14) for a single mode of type $(q_x, 0)$ if we make use of $A' = A(1 + \frac{1}{4}q_x^2 \, a_{q_x,0}^2)$. The negative sign in front of the correction term in κ' indicates the softening in this case.

An amusing illustration of the effect of parallel undulations is a rather stiff polymer closed to form a ring. Its thermal undulations in terms of curvature are not affected by the uniform bend and vice versa. Only the decrease of the ring radius in the presence of undulations signals a lowering of the effective bending stiffness of the polymer.

Orthogonal ripple: Ripples orthogonal to the cylindrical base curvature stiffen the membrane, in contrast to parallel ones. This is due to the fact that at the

crests of such a ripple the extra curvature ξ is of the same sign as the cylindrical curvature, and at the same time the membrane area is increased as compared to the background shape. In linear approximations the two effects obey

$$J'^2 = \frac{1}{r^2} - 2\frac{1}{r}\left(\frac{1}{r^2} + \partial_{zz}\right)\nu \; + \; O(\nu^2)$$

and

$$\sqrt{g'} = 1 + \frac{\nu}{r} \; + \; O(\nu^2) \; .$$

Calculating from these two expressions the total bending energy of a membrane with a single excited mode of wave vector $(0, q_z)$, we find the renormalized bending rigidity of the cylinder to be

$$\kappa' = \kappa(1 - (-q_z^2)\, a_{0,q_z}^2). \tag{18}$$

According to this formula the orthogonal ripple stiffens the membrane. It is four times as effective as a softening, parallel ripple of equal wavelength.

There is a second correction quadratic in ν to the bending energy due to the coupling between background curvature and orthogonal ripple. It originates from the sloped regions of the ripple where the membrane partially escapes cylindrical curvature. The reduced principal curvature is $(\cos\varphi)/r = (1/r)(1 - \varphi^2/2 + O(\varphi^4))$, φ being the slope angle. For a single mode of type $(0, q_z)$ the second effect and the increase of membrane area due to the slope result in a softening given by

$$\kappa' = \kappa(1 - \frac{1}{4}q_z^2\, a_{0,q_z}^2) \; . \tag{19}$$

Combining the opposite rigidity corrections of (18) and (19) results in a stiffening which agrees with (14).

At this point let us briefly consider what would be different in a derivation of κ' based on the displacement measure. Keeping ν fixed instead of ξ while the background is cylindrically bent implies that the regular part of the bending energy

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density is $1/2\ \kappa(\Delta\nu)^2$ instead of $1/2\ \kappa(\Delta\nu - \nu/r^2)^2$. Accordingly, the single mode Hamiltonian (14) would have to be recast in the form

$$
\int \frac{1}{2}\kappa J'^2 \sqrt{g'}\mathrm{d}x\mathrm{d}z =
$$
$$
\int \frac{1}{2}\kappa\left(\frac{1}{r^2} + (\vec{q}\,^2)^2\nu^2 + \frac{1}{r^2}(-\frac{5}{2}q_x^2 - \frac{1}{2}q_z^2 + O(r^{-2}))\nu^2\right)\mathrm{d}x\mathrm{d}z\ ,
$$

if again the third term is to represent the coupling of the undulations to cylindrical background curvature. The negative signs before q_x^2 and q_z^2 in the coupling term suggest that both parallel and perpendicular ripples soften the membrane. Absorbing the third term into the first renormalizes κ. The same manipulations as above, including thermal averaging and summation over all modes, would lead to

$$
\kappa' = \kappa - 3\frac{kT}{8\pi}\log M\ .
$$

This is the result obtained by all other authors. They generally used the displacement measure and took the approach via undulation mode entropies.

The undulations of unstretchable fluid membranes must be accompanied by lateral displacements. They can be analyzed mode by mode and consist of local and global parts. The local part redistributes membrane material between the crests and troughs of a ripple in a bent surface and in general varies linearly with the mode amplitude. The global part takes account of the area absorbed by the undulation and goes with the square of the amplitude. We have disregarded lateral motion, with one exception: In the case of the wavy polymer it is automatically included by employing arc length as independent variable. Inspection shows that lateral displacement need not be taken into account in calculations of the effective bending rigidity of polymers and membranes in the usual approximation that is quadratic in the mode amplitudes .

Renormalization of κ, c_s and $\bar{\kappa}$ via undulation mode entropies

The effective bending rigidity has been derived above for uniform cylindrical curvature and earlier for the spherical membrane, both times in the "frozen mode" approach. The same stiffening was obtained in both calculations. Let us try now to extend the result to more general geometries, showing simultaneously that the undulations do not affect the spontaneous curvature c_s and the modulus of Gaussian curvature, $\bar{\kappa}$, of the membrane. Obviously, we have to limit ourselves to "compact" shapes such as squares, circles and spheres. For the following calculations it is advantageous to follow the approach based on fluctuation mode entropies. The curvature increment $\xi = J' - J$ will again be used as measure of integration.

In order to calculate the free energy of the fluctuating membrane and to renormalize its elastic parameters, we employ a field–theoretic approach as has been introduced by Förster [7] in this frame. Considering some background configuration with parametrization $\vec{X}(\bar{\sigma})$, which describes the effective shape, we expand the bending Hamiltonian in (2) to second order around $\vec{X}(\bar{\sigma})$. We drop linear terms, assuming the background to be an equilibrium shape. The free energy F of $\vec{X}$ may then be written as [7, 10, 22]

$$F(\vec{X}) = H(\vec{X}) + \frac{1}{2}kT \log \det \frac{\delta^2 H(\xi)}{\delta\xi(\sigma)\delta\xi(\sigma')} + O((kT)^2) \ , \tag{20}$$

where ξ parametrizes the configurations $\vec{X}'$ in the vicinity of $\vec{X}$. After expanding the r.h.s. in powers of background curvature one may introduce effective elastic parameters marked by a prime, such as κ', and an effective Hamiltonian H' employing the new parameters in conjunction with J and K of the background shape $\vec{X}$.

To perform this program we first have to calculate the bending energies of the fluctuating membrane. Starting from eq. (7) we obtain

$$J'^2 = J^2 + 2J(D^2 + J^2 - 2K)\nu + ((D^2 + J^2 - 2K)\nu)^2$$

$$-J^2\ (D\nu)^2 + 2JJ^{ij}\ D_i\nu D_j\nu + 2J(J^3 - 3JK)\ \nu^2$$

$$+4JJ^{ij}\ \nu D_i D_j\nu + 2J\nu\ D_n\nu\ g^{ij}(D_j J_i^n + J\ \Gamma_{ij}^n) + O(\nu^3)\ . \tag{21}$$

Combining this with (6) yields

$$J'^2\sqrt{g'} = [J^2 - J^3\nu + 2J(D^2 + J^2 - 2K)\nu - \frac{1}{2}J^2(D\nu)^2$$

$$-2J^2\nu(D^2 + J^2 - 2K)\nu$$

$$+4JJ^{ij}\nu D_i D_j\nu + 2JJ^{ij}D_i\nu D_j\nu + ((D^2 + J^2 - 2K)\nu)^2$$

$$+JK\nu^2 + 2J(J^3 - 3JK)\nu^2 + 2J\nu D_n\nu g^{ij}(D_j J_i^n + J\Gamma_{ij}^n) + O(\nu^3)]\sqrt{g}\ . \tag{22}$$

We also need

$$J'\sqrt{g'} = [J + (D^2 + J^2 - 2K)\nu - J^2\nu$$

$$-J\nu(D^2 + J^2 - 2K)\nu + 2J^{ij}\nu D_i D_j\nu + J^{ij}D_i\nu D_j\nu$$

$$+JK\nu^2 + (J^3 - 3JK)\nu^2 + \nu D_n\nu g^{ij}(D_j J_i^n + J\Gamma_{ij}^n) + O(\nu^3)]\sqrt{g}\ . \tag{23}$$

In the following the background curvatures J and K are assumed to be weak and slowly varying. For all wave vectors except for the smallest ones the predominant terms quadratic in ν are those that contain at least two derivatives acting on ν. In calculating the effect of the short wavelength fluctuations upon the long distance elastic behaviour of membranes comprising a large number of molecules, it is sufficient to take into account only the latter. Giving rise to cutoff dependent effects, they are the only ones that can contribute to the renormalization of the elastic parameters. To replace the normal displacement ν by ξ we need the inversion of (4)

$$\nu = D^{-2}(1 + (J^2 - 2K)D^{-2})^{-1}\xi\ , \tag{24}$$

In the spirit of the above simplification, (24) can be approximated for terms which are quadratic in ν at the stage of J' and $g'^{1/2}$ (see eqs. (6) and (7)) by

$$\nu = D^{-2}\xi\ .$$

Disregarding irrelevant terms, we may thus write the bending energy as

$$
\int \mathrm{d}^2\sigma \ \sqrt{g'}(\frac{\kappa}{2}J'^2 + c_s\kappa J') = \int \mathrm{d}^2\sigma \ \sqrt{g}[\frac{\kappa}{2} \cdot (J^2
$$
$$
+2J\xi - J^3 D^{-2}(1 + (J^2 - 2K)D^{-2})^{-1}\xi
$$
$$
+\xi(\frac{1}{2}J^2 D^i D_i D^{-4} + 2JJ^{ij}D_iD_jD^{-4} - 2J^2D^{-2} + I)\xi
$$
$$
+\kappa c_s \cdot (J + \xi + \xi(-JD^{-2} + J^{ij}D_iD_j/D^4)\xi) + \text{irrel. terms}] \ , \tag{25}
$$

where we have performed some partial integrations, assuming either periodic bound-
ary conditions or undulations limited to a patch of membrane.

We are now in a position to set up the functional integral sweeping curvature space
that gives the effective free energy F of the background shape $\vec{X}$ with fields J and
K. In mechanical equilibrium linear terms in (25) drop out so that we may write

$$
F = -kT \log \int \mathcal{D}[\xi] \exp\left[-\frac{1}{kT} \int \mathrm{d}^2\sigma\sqrt{g} \left(\frac{\kappa}{2}J^2 - \kappa \ c_s J \right. \right.
$$
$$
+\frac{\kappa}{2}\left(\xi \left(1 + \frac{J^2}{2}D^{-2} + \frac{2JJ^{ij}D_iD_j}{D^4} \right. \right.
$$
$$
\left.\left.\left.\left. -\frac{2J^2}{D^2} + 2c_s\frac{J^{ij}D_iD_j - JD^2}{D^4} + \ \text{irrel. terms} \right) \xi \right) \right) \right] .\tag{26}
$$

Extracting the background energy and integrating the multiple Gaussian we are
led to [22]

$$
F = \int \mathrm{d}^2\sigma \ \sqrt{g} \ \left(\frac{\kappa}{2}J^2 - \kappa \ c_s J \right)
$$
$$
+\frac{kT}{2}\mathrm{Tr}'\log\left[\frac{\kappa}{2kT} \left(I + \frac{J^2}{2D^2} + \frac{2JJ^{ij}D_iD_j}{D^4} - \frac{2J^2}{D^2} + 2c_s\frac{J^{ij}D_iD_j - JD^2}{D^4} \right) \right] ,
$$
$$
\tag{27}
$$

where I is the identical operator. The prime on Tr indicates exclusion of some
modes at the low wave number cutoff. We assume rather weak background bends,
such that

$$
J^2, K \ <\simeq \ 1/L^2 \ , \tag{28}
$$

and may thus expand the logarithm in powers of them. A further limitation of J^2 may arise from c_s. The traces occuring in (27) are known to be [10, 23]:

$$\mathrm{Tr}' \frac{J^2}{D^2} \simeq -\int \mathrm{d}^2\sigma \ \sqrt{g} \ J^2 \cdot \frac{2}{4\pi} \log \frac{L}{a} \qquad \text{and} \tag{29}$$

$$\mathrm{Tr}' \frac{2JJ^{ij} \ D_i D_j}{D^4} \simeq -\int \mathrm{d}^2\sigma \ \sqrt{g} \ J^2 \cdot \frac{2}{4\pi} \log \frac{L}{a} \ , \tag{30}$$

L and a being upper and lower cutoffs due to finite membrane size and molecular distance, respectively. They are easily calculated for uniformly bent shapes such as squares, spheres and cylinders. One obtains

$$F = \int \mathrm{d}^2\sigma \ \sqrt{g} \ \frac{1}{2}\kappa J^2 \left(1 + \frac{kT}{4\pi\kappa} \log \frac{L}{a}\right) + \frac{kT}{2} \mathrm{Tr}' \log\left(\frac{\kappa}{2kT} I\right)$$
$$- \int \mathrm{d}^2\sigma \ \sqrt{g} \ c_s \kappa J \cdot \left(1 + \frac{kT}{4\pi\kappa} \log \frac{L}{a}\right) \ . \tag{31}$$

The middle term of (31) is equal to the free energy of the corresponding flat reference membrane. After subtracting it we are left with the free energy of bending which may be written in the renormalized form

$$\Gamma = \int \mathrm{d}^2\sigma \ \sqrt{g} \ (\frac{1}{2}\kappa' J^2 - \kappa' c_s' J) \ , \tag{32}$$

where

$$\kappa' = \kappa + \frac{kT}{4\pi} \log \frac{L}{a} \tag{33}$$

$$c_s' = c_s \ . \tag{34}$$

For comparison with (1), note that $L^2/a^2 = M$. Evidently, the effective bending rigidity comes out exactly as calculated for the sphere [13] and the cylinder, while the spontaneous curvature is not modified by the thermal undulations.

Formula (32) omits the Gaussian curvature term of the bending energy. We dropped it in the unrenormalized bending energy since, as a consequence of the Gauss–Bonnet theorem, the variation of Gaussian curvature does not affect its integral if the membrane is closed or satisfies periodic boundary conditions. Nevertheless, an

additional Gaussian curvature term could have emerged in the calculation of the effective bending rigidity, as it does when normal displacement is the measure of integration [18]. From the fact that such a term is absent in eq. (31) we may infer that $\bar{\kappa}$ is not affected by thermal undulations, i.e.

$$\bar{\kappa}' \; = \; \bar{\kappa} \; , \tag{35}$$

in agreement with previous considerations [13].

In the preceding calculation of the effective bending rigidity we did not specify the shape of the membrane, except for stipulating that it be "compact". Although the formula for κ' seems to be rather robust, because of the logarithmic dependence on wave vector cutoffs, it is not expected to hold for filamentous membranes. Specifically, the bending rigidity of the membrane closed to form a cylinder will not stiffen indefinitely as the cylinder is made longer. This is why we chose periodic boundary conditions with an axial period as long as the circumference in our calculation for the cylinder.

The renormalized elastic parameters calculated above are independent of the base curvatures J and K. It is an open question to which extent they remain valid if these curvatures are nonuniform over the base area. In the case of a wavy background one commonly uses as upper limit in (31) its wavelength or a fraction thereof instead of the membrane size L. This is because ripples of larger wavelength are part of the immutable background curvature.

Conclusion

We have renormalized the bending elastic parameters of fluctuating fluid membranes for cylindrical and more general membrane shapes. The same logarithmic increase of the bending rigidity with membrane size was obtained as previously for spherical vesicles. In addition, the modulus of Gaussian curvature and, for the first time,

spontaneous curvature were shown not to be affected to lowest order by thermal undulations. Apart from the illustrative treatment of the cylindrical membrane, we used arbitrary curvilinear coordinates in the calculations to obtain generally valid results as far as possible.

The predicted stiffening of fluid membranes by their thermal undulations may seem contrary to intuition, although there is no conflict with de Gennes' [2] derivation of the persistence length of membrane orientation. In the case of polymers the loss of orientational correlation unavoidably results in a breakdown of bending elasticity. This could be different in the case of membranes because of their two-dimensional connectedness and, in particular, the well-known scale invariance of bending energies. Moreover, it remains to be checked if the stiffening survives at very low bending rigidities ($\kappa \approx kT$), where the nearly flat approximation is inapplicable at any scale.
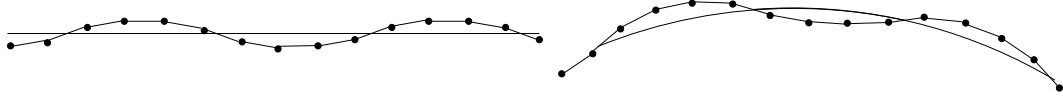
Acknowledgement

Figures

Figure 1: Wavy polymer with fixed ripple curvature amplitude on flat and bent background, respectively. Note the enhanced normal displacement from the background on the right–hand side.
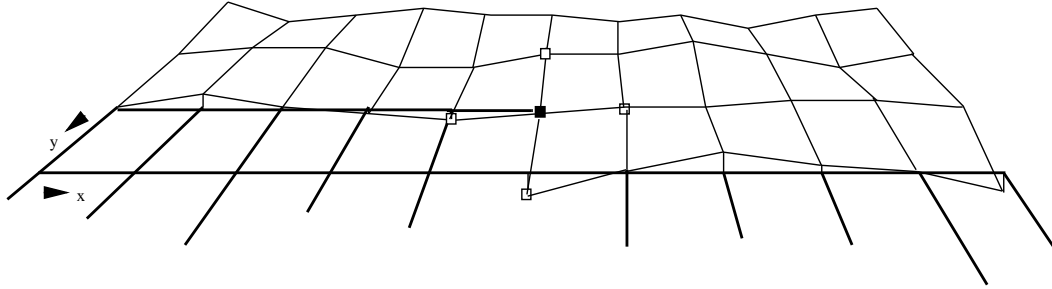

Figure 2: Lattice model of a membrane fluctuating around a flat base. The heavy lines are grid lines and shown only up to the membrane edge. The thin lines indicate membrane shape or, at membrane edge, normal displacement from the base. The five square-shaped points determine the mean curvature in the middle (solid square) of the plaquette

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